

New determination of the Ba-Mo yield matrix for ^{252}Cf S.-C. Wu,* R. Donangelo,[†] and J. O. Rasmussen*Lawrence Berkeley National Laboratory, Berkeley, California 94720*A. V. Daniel,[‡] J. K. Hwang, A. V. Ramayya, and J. H. Hamilton*Department of Physics and Astronomy, Vanderbilt University, Nashville, Tennessee 37235*

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Using triple-coincidence events of prompt fission gamma rays from 1995 Gammasphere data on spontaneous fission of ^{252}Cf , we made a careful analysis of the yield matrix of coincident pairs of barium ($Z=56$) and molybdenum ($Z=42$) fission fragments. Branching from gamma bands ($K=2$) and octupole bands ($K=0$) are presented for even-even partners, where observable. From this reanalysis the previously proposed “extra-hot-fission mode” (up to ten neutrons evaporated) as determined by twofold coincidences of 1993 Gammasphere data is much weaker, but not excluded. This finding is in agreement with a recently published similar study from the Legnaro gamma-detector array.

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Recent years have seen great advances in the study of prompt γ rays accompanying spontaneous fission. Although such research has progressed significantly since the availability of high-resolution germanium detectors around 35 years ago, the completion of large detector arrays like Gammasphere and Eurogam has produced a new spurt of interest in this field [1]. Ter-Akopian and collaborators have done pioneering work on the quantitative determination of yield matrices, using γ - γ coincidence data to extract yields of particular fission fragment pairs [2–4]. By simple arithmetic one finds the number of fission neutrons associated with each pair of coincident fragments. The yield of so-called “cold fission” events, where no neutrons are emitted, can be studied [3], complementing earlier studies with neutron detectors and providing an increased sensitivity [5]. Another interesting finding was that $\approx 14\%$ of the ^{252}Cf barium-molybdenum split goes via a “hot fission” mode, where as many as 10 neutrons are emitted [1,4]. This latter feature has stimulated some theoretical speculations and also some skepticism, since the hot fission mode (called Mode 2) has only been seen in the Ba-Mo pairs in ^{252}Cf and not in ^{248}Cm spontaneous fission [6]. There have been some theoretical efforts to understand how this hot fission could arise [4,7,8].

In the present work we used the 1995 Gammasphere data, taken by the GANDS95 Collaboration [9]. The analysis is carried out with the uncompressed triple coincidence spectra. This differs from the previous analyses where either uncompressed double coincidence spectra or compressed triple coincidence spectra were used. In the triple coincidence spectra, the compression factor varies from 2 to 3 channels per bin in the 300-keV region to as much as 8 channels per bin in the 2-MeV energy region [10]. In both of these methods one

faces problems, because of the vast number of γ rays in the spectrum. If one analyzes the twofold coincidence spectra, the problem is a large background and considerable chance of unrelated coincident gamma pairs overlapping. With compressed triple coincidence data, the chance of accidental overlap in three-dimensional (3D) space is less, but compression makes it impossible to resolve close-lying peaks by peak-fitting routines.

We wish to point out here in our revised manuscript that we were unaware of the prior submission of similar conclusions from the Legnaro gamma detector array published this year by Biswas *et al.* [11]. They also show a yield discrepancy with Ter-Akopian *et al.* regarding the anomalous Mode 2, finding it much weaker than originally reported but not excluded. Our results have been presented elsewhere [12].

In this new analysis, using the uncompressed 3D data, we measured anew the pair yields of barium ($Z=56$) with molybdenum ($Z=42$) partners. Because the ^{104}Mo and ^{108}Mo have $2^+ \rightarrow 0^+$ transitions too close in energy to resolve and $4^+ \rightarrow 2^+$ transitions that are barely resolvable with peak-fitting routines, we have generally chosen to double-gate on the Ba fragments and measure the $2^+ \rightarrow 0^+$ intensities in the Mo partners (and $4^+ \rightarrow 2^+$ where the $2^+ \rightarrow 0^+$ are unresolvable). We also measured the 2^+_{γ} -to-ground intensities for information on relative population of the $K=2$ band arising from the triaxiality of the Mo fragments [13–16]. The barium double gates are on the $4 \rightarrow 2 \rightarrow 0$ cascade and the $3 \rightarrow 2 \rightarrow 0$ cascade, the latter being significant in the heavier bariums where octupole deformation tendencies [1,17] are reported. The odd- A nuclei are special cases, and we shall discuss in a separate publication the details of their analysis, which depends on unique individual level schemes with different parallel feeding patterns. Their yields in our triple-coincidence analysis here fall rather smoothly into the yield patterns of their even-even neighbors, as will be seen in tables that follow. It is, of course, true that analysis of triple-gamma coincidence events cannot fully reproduce yields from analysis of two-fold coincidence events. Furthermore, the triple-gamma events need to be summed over all gates involving γ rays feeding the 2^+ first-excited states of the

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TABLE I. Pairwise percentage yields of Mo-Ba fission fragments. The numbers in parentheses after each value are the statistical standard deviations (s.d.). See text for further details.

Yield	¹³⁸ Ba	¹⁴⁰ Ba	¹⁴¹ Ba	¹⁴² Ba	¹⁴³ Ba	¹⁴⁴ Ba	¹⁴⁵ Ba	¹⁴⁶ Ba	¹⁴⁷ Ba	¹⁴⁸ Ba
¹⁰² Mo		<0.007		<0.008	0.033(11)	0.094(9)	0.174(8)	0.206(4)	0.053(6)	0.029(3)
¹⁰³ Mo		0.024(11)	0.041(19)	0.051(7)	0.176(18)	0.587(10)	0.698(20)	0.458(9)	0.181(9)	0.035(6)
¹⁰⁴ Mo	0.003(2)	0.058(9)	0.052(6)	0.23(1)	0.46(2)	1.250(11)	0.767(11)	0.457(6)	0.045(6)	0.008(2)
¹⁰⁵ Mo	<0.009	0.062(11)	0.116(15)	0.555(12)	0.887(25)	1.280(15)	0.532(22)	0.135(8)	<0.009	
¹⁰⁶ Mo	0.004(2)	0.136(7)	0.197(11)	0.866(10)	0.825(17)	0.733(10)	0.141(8)	0.035(3)		
¹⁰⁷ Mo	0.018(3)	0.226(8)	0.285(18)	0.441(10)	0.308(20)	0.184(8)	0.034(8)			
¹⁰⁸ Mo	0.004(1)	0.189(8)	0.142(15)	0.137(8)	0.053(8)	0.015(5)				

double-gated partner. Indeed, we have summed over gamma feeding, not only from the 4^+ second-excited member of the ground band, but also from significant second 2^+ (gamma-band) states and 3^- (octupole) states. Obviously, fission events in which the 2^+ states of each fission partner were directly populated without any preceding γ rays emitted would never be recorded among triple-gamma coincidence events. It seems from general rotational band feeding patterns that the former class of events—with only two γ rays—will not be of significant intensities. In the ¹³⁸Ba yields any direct 2^+ population without preceding γ ray would be unmeasurable in these thick-source experiments because the 2^+ lifetime is much shorter than the fission-fragment stopping time, resulting in broad Doppler smearing. Yields involving ¹⁰⁴Mo direct population without preceding gammas would be unresolvable from ¹⁰⁸Mo population, since the $2 \rightarrow 0$ transition energies are unresolvably close. Therefore, the triple-coincidence analysis will not miss yields of direct feeding to 2^+ states of ¹³⁸Ba, ¹⁰⁴Mo, or ¹⁰⁸Mo, compared to the earlier work, since they are unmeasurable in double coincidence—only the sum of the two Mo isotopes would be measurable. In the yield calculations we have taken into account that Compton suppression is not complete and that, also, Compton scattering on the walls of the chamber and into a detector, as well as true continuum gammas are simultaneously present. Hence, some γ rays of energies higher than the gated gammas may be contributing to a given peak. In order to correct for this, on each double-gated spectrum we have taken two spectra with each gate successively shifted to a nearby low valley but away from the gate peak. Half of the sum of these background spectra is subtracted from the peak. The correction is usually negligible for strong peaks but often significant for weaker peaks. Spectral triple-coincidence counts are divided by the product of the three γ ray efficiencies and the product of the corrections for internal conversion. Rather than using one of the existing gamma efficiency curves for Gammasphere, as determined off-line with radioactive standards in singles mode, we checked the efficiency curves with rotational cascades in the actual experiment, double-gating on two transitions high in the rotational band and measuring the intensities of the lower transitions in the band. Thus, these efficiency measurements involved coincidence efficiencies and take into account Compton suppression, “time-walk,” and other factors at the high count rates of the actual experiment.

In Table I we give the fission yields in percent. The col-

umns are labeled by the Ba mass numbers and the rows by the Mo mass numbers. The numbers in parentheses after each value are the statistical standard deviations (s.d.), taken as the square root of the sum of the squares of the peak fit of the value and the average of the squares of the two shifted-gate background subtractions. These standard deviations are taken directly from Radford’s gf2 least-squares multiple-peak-fitting code (RADWARE). In cases where the resulting overall s.d. is greater than the value after background subtraction, we have entered only an upper limit ($<$) given by the value plus two standard deviations (approximately 95% confidence level). We realize that these reported statistical standard deviations do not take into account various systematic errors, and that therefore the effective overall uncertainties in the yields are significantly larger.

We have summed the $4^+ \rightarrow 2^+ \rightarrow 0^+$ cascade and the $3^- \rightarrow 2^+ \rightarrow 0^+$ cascade contributions of the Ba gates and the $2^+ \rightarrow 0^+$ and the $2_\gamma^+ \rightarrow 0^+$ Mo peaks in the resulting spectra to give yields in Table I as close as possible to two-fold coincidence yields. Our yields are normalized so that the sum of ¹⁰⁶Mo yields matches that of Ter-Akopian *et al.* [18]. Figure 1(a) is a logarithmic contour plot of our yields, and Fig. 1(b) is the corresponding logarithmic contour plot from the published data of Ter-Akopian *et al.* [18]. The innermost contour lies at the 1% fission yield, and the successive outward contours are spaced down by decades.

In Table II we show the yield of $2_\gamma \rightarrow 0$ relative to $2^+ \rightarrow 0^+$ in the Mo partners. In Table III we show the yield of the octupole-quadrupole $3^- \rightarrow 2^+ \rightarrow 0^+$ relative to the normal $4^+ \rightarrow 2^+ \rightarrow 0^+$ gating in the barium partners. The contour plots of yields do not show evidence for any highly excited mode in nearly the 14% abundance reported in the earlier measurements [1,4]. We note the peculiarities that make ¹³⁸Ba especially difficult to measure in two-fold coincidence analysis. The $6^+ \rightarrow 4^+$ transition in neutron-closed-shell ¹³⁸Ba is almost identical in energy (191.96 keV) to the ¹⁰⁴Mo and ¹⁰⁸Mo $2^+ \rightarrow 0^+$ transitions, 192.2 and 192.8 keV, respectively. Furthermore, the 6^+ level is fed by β decay from the higher-spin ¹³⁸Cs isomer in addition to possible direct fission population. Ter-Akopian *et al.* [18] took into account the β -decay feed in their 1997 paper, and reduced their earlier reported yield by about 50%. As discussed earlier in this paper, the half-life of the first excited 2^+ state in ¹³⁸Ba, 0.192 ps, is so much shorter than the stopping time of fission fragments in the surrounding nickel foils that any

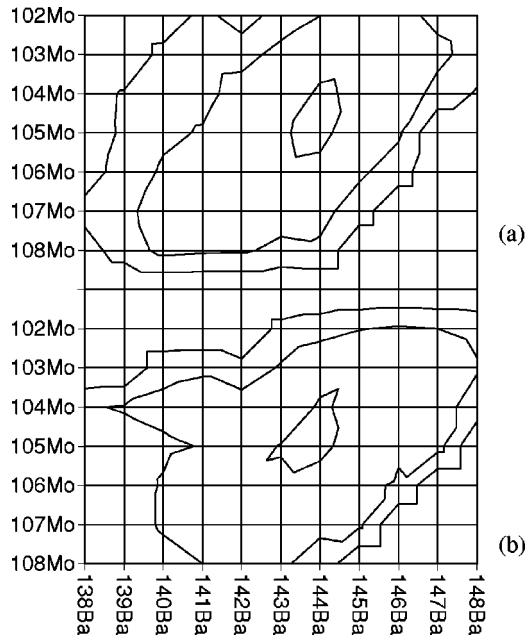


FIG. 1. Logarithmic contour plots of the Ba-Mo yield matrix. (a) is the new analysis of this paper using triple-coincidence data, and (b) is based on the published data of Ter-Akopian *et al.* [16]. In both cases to facilitate contour plotting we have used interpolated yields for the unmeasured ^{139}Ba , as the geometric mean of yields for neighboring ^{138}Ba and ^{140}Ba . All yields are normalized to absolute percent yield per spontaneous fission (see text). The central contour at the peak is the 1% level and successive outer contours go down by factors of 10. Unmeasured yields, low or zero, outside the data blocks have been represented as 0.001% to avoid forming spurious contour lines for smaller limiting values. Irregularities in the 0.001% contour line are not to be taken seriously, since few measured points are that low.

direct prompt fission feed of the 2^+ state would experience so much Doppler smearing as to be unobservable. Also any prompt fission feed of the 6^+ state would not show coincidences with Mo fragment gamma spectra, since the 6^+ half-life of 800 ns is considerably longer than the 200 ns time resolution with which the 3D histogram was constructed. It is possible to look for prompt fission gamma coincidences with Mo partners only by using the $4^+ \rightarrow 2^+ \rightarrow 0^+$ cascade in ^{138}Ba and some Mo transition other than the 192-keV line because the 4^+ half-life is 2.17 ns, well within the limits of the 200 ns resolving time and the few-picosecond stopping time. It is by using this cascade that we have set the ^{138}Ba yields given in Table I, though it must be borne in mind that only direct fission feed through the 4^+ state was measurable

TABLE II. Relative yield from Mo gamma band. The numbers in parentheses after each value are the statistical standard deviations (s.d.). See text for further details.

Neutron loss \rightarrow	-4	-2
^{102}Mo		
^{104}Mo	0.027(3)	0.038(6)
^{106}Mo	0.071(8)	0.072(10)

TABLE III. Pair yield of Mo $3 \rightarrow 2 \rightarrow 0$ relative to $4 \rightarrow 2 \rightarrow 0$. The numbers in parentheses after each value are the statistical standard deviations (s.d.). See text for further details.

Neutron loss \rightarrow	-6	-4	-2	0
^{102}Mo	0.050(17)	0.117(10)		
^{104}Mo		0.123(5)	0.169(7)	
^{106}Mo			0.046(3)	0.281(55)

in our thick-source experiment. Figures 1(a) and 1(b) of the yield matrix show a maximum for ^{104}Mo and ^{105}Mo with a ^{144}Ba partner, a neutron loss of 4 and 3, respectively. These new data do not show evidence for a westward spur that would correspond to the Mode 2 of extra high excitation energy. The only significant difference between the published double-coincidence analysis and our new triple-coincidence analysis is in the ratios between ^{104}Mo and ^{108}Mo yields with partners of $^{138,140,141}\text{Ba}$. We believe our resolving of the $4^+ \rightarrow 2^+$ transitions of the above Mo isotopes gives the most reliable ratio. In this we carefully checked the possibility of ^{108}Mo being too heavily weighted by virtue of the closeness in energy of three of the rotational band transitions in ^{108}Mo and ^{140}Ba (the 527.2 keV $6^+ \rightarrow 4^+$ in the former and the 529.7 keV $8^+ \rightarrow 6^+$ and 528.25 keV $6^+ \rightarrow 4^+$ in the latter). Our check consisted of setting one gate on the 602.35 keV $2^+ \rightarrow 0^+$ of ^{140}Ba and sliding the narrowest one-channel second gate over the region of the above three close-lying transitions. The examination of the line shape in the spectrum of the $4^+ \rightarrow 2^+$ transitions of ^{104}Mo and ^{108}Mo showed little change, the ^{104}Mo line remaining a tiny shoulder on the low side of the ^{108}Mo line. The triple-coincidence yields of this paper do not show the strong northwesterly spur of “Mode 2” on the contour map of Fig. 1(b) but are otherwise in good agreement.

Turning to the population of side bands, what can we conclude from Tables II and III? One might guess that the

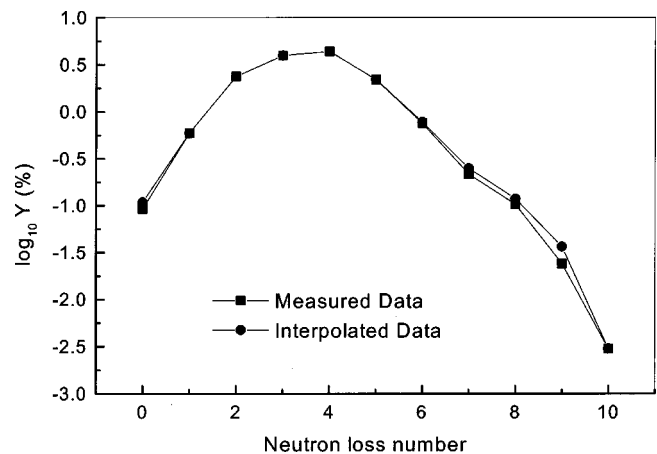


FIG. 2. Semilog plot of summed Ba-Mo fission yields vs neutron-loss number. The lower curve uses just measured values of Table I, while the higher curve adds in logarithmically interpolated values, where measured values were on either side of the missing yield.

relative population of higher bands associated with pear-shaped or triaxiality tendencies would increase with degree of collectivity. There is some trend of relative population with the energy of the collective excited state in the sense expected by a thermal model. However, it is hard to draw firm conclusions yet on systematics of side-band populations. Of course, to compare our new triple-coincidence analysis with twofold coincidence work it was necessary to measure and add in side-band feeding to first-excited states.

A more quantitative plot for evaluating possibilities of a hot fission mode is Fig. 2. There the sum of yields for a particular number of neutrons lost is plotted. The lower curve is a sum of measured yields of Table I, excluding cells with only upper limits determined. The upper curve fills missing cells with adjacent measured yields, mainly ^{139}Ba , not measured. The interpolated values are the geometric mean of the adjacent cell values. Both curves show a hint of a shoulder around eight neutrons lost. In the work of Biswas *et al.* [11] their analogous plot also shows a similar small irregularity around eight neutrons lost. They reported they could not observe a ten-neutron loss. We do report one such

cell, ^{104}Mo - ^{138}Ba , but with a large standard deviation as $0.003 \pm 0.002\%$. The evidence calling for further study of the possible hot fission “shoulder” stands with or without the ten-neutron-loss yield.

As to our differences with the prior two-fold coincidence studies, we must emphasize the great challenges posed by the overly rich γ -ray forest of prompt and β -delayed fission γ rays.

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